## **PCT**

#### WORLD INTELLECTUAL PROPERTY ORGANIZATION International Bureau



## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification 7:

B01J 19/10

(11) International Publication Number:

WO 00/35579

A1

(43) International Publication Date:

PT, SE).

22 June 2000 (22.06.00)

(21) International Application Number:

PCT/GB99/03857

(22) International Filing Date:

22 November 1999 (22.11.99)

(30) Priority Data:

9827360.0

12 December 1998 (12.12.98) GB **Published** 

With international search report.

(81) Designated States: GB, JP, US, European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL,

(71) Applicant (for all designated States except US): AEA TECH-NOLOGY PLC [GB/GB]; 329 Harwell, Didcot, Oxfordshire OX011 ORA (GB).

(72) Inventor; and

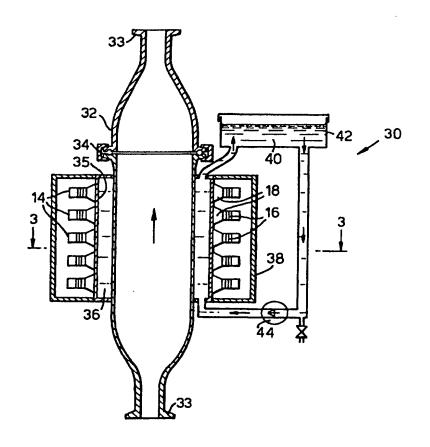
(75) Inventor/Applicant (for US only): PERKINS, John, Patrick [GB/GB]; Sonic Systems, Bakers Farm Barns, Puckington, Ilminster, Somerset TA19 9JA (GB).

(74) Agents: MANSFIELD, Peter, Turquand et al.; AEA Technology plc, Patents Dept., 329 Harwell, Didcot, Oxfordshire OX11 ORA (GB).

(54) Title: PROCESS AND APPARATUS FOR IRRADIATING FLUIDS

#### (57) Abstract

Fluids in a vessel are subjected to a high ultrasonic intensity, by means of several ultrasonic transducers attached to a wall of the vessel, each transducer (14) radiating no more than 3 W/cm<sup>2</sup>, the transducers being sufficiently close to each other, and the number of transducers being sufficiently high, that the power dissipation within the vessel is at least 25 W/litre. The number of transducers (14), the power of the transducers, and the volume of the vessel may be such that the power density is between 40 and 80 W/litre. The vessel may be double walled, and the space between the two walls be filled by a low attenuation buffer liquid (36) whose cavitation threshold is above that of the liquid to be treated.



### FOR THE PURPOSES OF INFORMATION ONLY

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AL	Albania	ES	Spain	LS	Lesotho	SI	Slovenia
AM	Armenia	FI	Finland	LT	Lithuania	SK	Slovakia
ΑT	Austria	FR	France	LU	Luxembourg	SN	Senegal
ΑU	Australia	GA	Gabon	LV	Latvia	SZ	Swaziland
AZ	Azerbaijan	GB	United Kingdom	MC	Monaco	TD	Chad
BA	Bosnia and Herzegovina	GE	Georgia	MD	Republic of Moldova	TG	Togo
вв	Barbados	GH	Ghana	MG	Madagascar	TJ	Tajikistan
BE	Belgium	GN	Guinea	MK	The former Yugoslav	TM	Turkmenistan
BF	Burkina Faso	GR	Greece		Republic of Macedonia	TR	Turkey
BG	Bulgaria	HU	Hungary	ML	Mali	TT	Trinidad and Tobago
ВЈ	Benin	ΙE	Ireland	MN	Mongolia	UA	Ukraine
BR	Brazil	IL	Israel	MR	Mauritania	UG	Uganda
BY	Belarus	IS	Iceland	MW	Malawi	US	United States of America
CA	Canada	ľT	Italy	MX	Mexico	UZ	Uzbekistan
CF	Central African Republic	JP	Japan	NE	Niger	VN	Viet Nam
CG	Congo	KE	Kenya	NL	Netherlands	YU	Yugoslavia
CH	Switzerland	KG	Kyrgyzstan	NO	Norway	zw	Zimbabwe
CI	Côte d'Ivoire	KP	Democratic People's	NZ	New Zealand		
CM	Cameroon		Republic of Korea	PL	Poland		
CN	China	KR	Republic of Korea	PT	Portugal		
CU	Cuba	KZ	Kazakstan	RO	Romania		
CZ	Czech Republic	LC	Saint Lucia	RU	Russian Federation		
DE	Germany	LI	Liechtenstein	SD	Sudan		
DK	Denmark	LK	Sri Lanka	SE	Sweden		
EE	Estonia	LR	Liberia	SG	Singapore		

- 1 -

#### Process and Apparatus for Irradiating Fluids

The present invention relates to a process and an apparatus for subjecting fluids to irradiation by high intensity ultrasound.

If a liquid is subjected to a high ultrasonic intensity, that is to say an intensity greater than about 0.3 W/cm² at a frequency typically in the range 10 kHz to 100 kHz, then there is a significant deposition of energy into the liquid through attenuation and non-linear effects. This may lead to physical changes (for example streaming, mixing, or emulsification) or to chemical changes, and the present invention is principally concerned with the latter, which may be referred to as sonochemistry. The most significant sonochemical effects are usually associated with cavitation in the liquid, especially transient vaporous cavitation, which typically only occurs at an ultrasonic intensity above a threshold which is typically above 0.3 W/cm² and is different for different liquids.

An apparatus for subjecting a liquid to a very high intensity is described in GB 2 243 092 B (UKAEA), in

25 which a transducer is attached to one end of a tapered resonant coupler, the coupler having a nodal flange; this flange is attached to a sleeve projecting from the wall of a pipe containing the liquid, and the space around the sides and end of the coupler within the sleeve is filled with a coupling liquid such as olive oil. The use of such a nodal flange mounting enables a transducer of higher power to be used, but the apparatus is somewhat complex. It has also been suggested, for example in US 4 433 916

- 2 -

(Hall), to attach a plurality of acoustic transducers directly to the wall of a cylindrical vessel containing a fluid, with a specified phase difference between the acoustic signals applied to adjacent transducers.

5 Similarly, US 4 369 100 (Sawyer) describes attaching ultrasonic transducers to the outer wall of an annular duct so that both the inner and outer walls are set in resonance. And US 2 578 505 (Carlin) also describes a pipe with several transducers attached to the surface 10 arranged in one or more circumferential rings.

According to the present invention there is provided an apparatus for subjecting a fluid to irradiation by high intensity ultrasound, the apparatus comprising a vessel, and a plurality of ultrasonic transducers attached to a wall of the vessel so as to radiate ultrasonic waves into a fluid in the vessel, the vessel being large enough that each transducer radiates into fluid at least 0.1 m thick, each transducer radiating no more than 3 W/cm<sup>2</sup>, the transducers being sufficiently close to each other, and the number of transducers being sufficiently high, that the power dissipation within the vessel is at least 25 W/litre but no more than 150 W/litre.

25

The values of power given here are those of the electrical power delivered to the transducer or the transducers, as this is relatively easy to determine. There will inevitably be losses in converting electrical to acoustic power, and in transmitting the acoustic power from the transducer into the fluid within the vessel, but these are difficult to assess accurately. The transducer

- 3 -

typically is at least 90% efficient in converting electrical to acoustic power.

The power radiated by each transducer may for

5 example be in the range 1-2 W/cm<sup>2</sup>. This is a similar power intensity to that used in ultrasonic cleaning baths, and is above the threshold required to achieve cavitation. The resonant ultrasonic coupler with a nodal flange (as described in GB 2 243 092 B) can achieve significantly higher power intensities, for example radiating more than 10 W/cm<sup>2</sup>, but the lower intensity ultrasound generated by the present invention can propagate much further through a liquid, and the stresses in the transducers are much less.

15

The thickness of fluid into which each transducer radiates, that is the width or diameter of the vessel, is preferably no more than 1.2 m, and in a preferred embodiment the vessel is cylindrical of diameter in the range 0.2 to 0.6 m, such as 0.3 m.

Preferably the number of transducers, the power of the transducers, and the volume of the vessel are such that the power density is between 40 and 80 W/litre, more preferably between 60 and 70 W/litre. Although this does not produce such high power densities as can be achieved with an irradiator as described in GB 2 243 092 (UKAEA), it enables a larger volume to be irradiated and with greater uniformity, at sufficient intensity to cause cavitation; this is partly because very high cavitation levels near the vessel wall can have the effect of shielding parts of the fluid further from the wall.

- 4 -

In a preferred embodiment the vessel is double walled, the transducers being attached to the outer wall, the fluid to be treated being enclosed within the inner wall, and the space between the two walls being filled by a low attenuation buffer liquid whose cavitation threshold is above that of the liquid to be treated.

The width of the gap between the two walls is desirably equal to a quarter wavelength in the coupling liquid (which for a frequency of 20 kHz would be about 18 mm), or an odd-numbered multiple of that distance, to optimise the matching of impedance.

In one embodiment some of the transducers are
15 energized at one frequency and other transducers at a
different frequency, for example at 20 kHz and at 40 kHz.
This can be very effective at causing cavitation and
energy deposition within a fluid.

The invention will now be further and more particularly described, by way of example only, and with reference to the accompanying drawings in which:

Figure 1 shows a longitudinal sectional view through an 25 irradiator apparatus;

Figure 2 shows a longitudinal sectional view through an alternative irradiator apparatus; and

30 Figure 3 shows a sectional view on the line 3-3 of figure 2.

- 5 -

Referring to figure 1 an irradiator 10 incorporates a stainless-steel duct 12 of internal diameter 0.31 m and of wall thickness 2 mm. To the outside of the wall are attached sixty transducer modules 14 closely packed in a square array. Each transducer module 14 comprises a 50 W piezoelectric transducer 16 which resonates at 20 kHz, attached to a conically flared titanium coupling block 18 by which it is connected to the wall, the wider end of each block being of diameter 63 mm. The transducers 10 modules 14 are arranged in five circumferential rings each of twelve modules 14, the centres of the coupling blocks 18 being on a square pitch of 82 mm. The irradiator 10 also incorporates five signal generators 20 (only one is shown) each of which drives all the 15 transducers 16 in a ring. All the transducers 16 are activated at 20 kHz.

In use of the irradiator 10, a liquid is caused to flow through the duct 12 and each transducer 16

20 activated. Each transducer 16 radiates 50 watts over a circle of diameter 63 mm, that is an intensity of 1.6 W/cm<sup>2</sup>. The energy from all the transducers 16 is dissipated over the cylindrical volume of the duct 12, that is to say over a volume of about 31 litres (that is the volume enclosed by the array of transducer modules 14), so the power density is about 97 W/litre, or about 80 W/litre if the ultrasonic irradiation is also effective 40 mm beyond each end of the array of transducer modules 14.

30

In one modification, three of the rings of transducers 16 are as described above, while the other two rings resonate at 40 kHz and are driven at that

frequency (the rings being of alternate frequency along the duct 12). The power intensity and power density are as described earlier, but the fluid is consequently exposed simultaneously to two different frequencies generating cavitation. This can produce more effective sonochemical results.

Referring now to figures 2 and 3 there is shown an alternative irradiator 30 which has many features in 10 common with that of figure 1. The irradiator 30 includes a generally cylindrical duct 32 of polytetrafluoroethylene (PTFE) of internal diameter 0.31 m and of wall thickness 3 mm, which tapers at each end down to an internal diameter of 0.10 m and a wall thickness of 6 mm 15 provided with a flange 33 for connection to other process ducts (not shown), and has a sealed joint 34 for inspection or cleaning purposes. Around the outside of the duct 32 is a concentric stainless-steel steel tube 35 of wall thickness 1 mm and of external diameter 0.354 m, such that there is a gap 36 of width 18 mm between the 20 duct 32 and the tube 35. Sixty transducer modules 14 are attached to the outer surface of the tube 35 in a rectangular array forming five rings of twelve, the spacing between the centres of the coupling blocks 18 25 being 82 mm parallel to the longitudinal axis of the tube 35 and 92.7 mm circumferentially. The array of transducers 14 is enclosed by an acoustic shield 38. coupling liquid such as olive oil 40 is used to fill the gap 36 and is re-circulated from a reservoir 42 by a pump 30 This coupling liquid has a higher threshold for cavitation than water, and has an impedance which is between that of the titanium coupling block 18 and that of the fluid within the duct 32 (typically mainly water,

- 7 -

or an industrial solvent); at a frequency of 20 kHz the wavelength of the sound in this oil is 72 mm, so that the gap 36 is of width equal to a quarter of the wavelength.

- In use of the irradiator 30, a liquid is caused to flow through the duct 32 and each transducer 16 is activated by a power supply (not shown) at 20 kHz. The dissipated power intensity and power density are as described in relation to figure 1. The impedance

  matching provided by the oil in the gap 36 allows more of the applied power to enter the fluid within the duct 32 while reducing erosion at the inner, irradiating, surface of the tube 35.
- 15 It will be appreciated that the power intensity in the irradiator 30 may be increased by increasing the numbers of transducer modules 14 in each ring. For example there might be sixteen transducer modules 14 in each ring, if the coupling blocks 18 were arranged on a circumferential spacing of 69.5 mm. This would increase both the power density and the power intensity by 33 percent. It will also be appreciated that the duct 32 can be of any material suited to the liquid being processed, and that it may form part of a pressure vessel. It will also be appreciated that the duct 12 or 32 need not be cylindrical but might for example be of square cross-section.

Furthermore the ultrasonic frequencies or frequencies might be different from those described above, and might be as high as 140 kHz or even 200 kHz, as such high frequencies tend to reduce the risk of cavitation erosion.

#### Claims

- An apparatus for subjecting a fluid to irradiation by high intensity ultrasound, the apparatus comprising a vessel (12, 32), and a plurality of ultrasonic transducers (14) attached to a wall of the vessel so as to radiate ultrasonic waves into a fluid in the vessel, characterised by the vessel (12, 32) being large enough that each transducer (14) radiates into fluid at least
   0.1 m thick, each transducer (14) radiating no more than 3 W/cm², the transducers (14) being sufficiently close to each other, and the number of transducers (14) being sufficiently high, that the power dissipation within the vessel (12, 32) is at least 25 W/litre but no more than
   150 W/litre.
  - 2. An apparatus as claimed in claim 1 wherein the power radiated by each transducer (14) is in the range 1-2  $\rm W/cm^2$ .

20

25

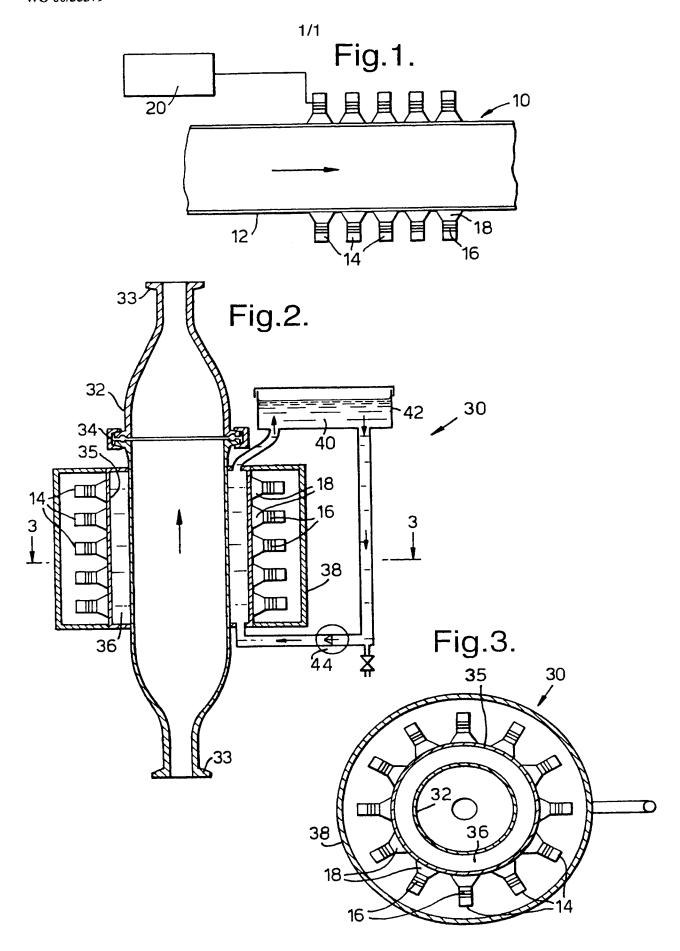
- 3. An apparatus as claimed in claim 1 or claim 2 wherein the number of transducers (14), the power of the transducers (14), and the volume of the vessel (12, 32) are such that the power density is between 40 and 80 W/litre.
- An apparatus as claimed in any one of the preceding claims wherein the vessel (32) is double walled, the transducers (14) being attached to the outer wall (35),
   the fluid to be treated is enclosed within the inner wall (32), and the space (36) between the two walls (32, 35) is filled by a low attenuation buffer liquid whose

- 9 -

cavitation threshold is above that of the liquid to be treated.

5. An apparatus as claimed in any one of the preceding claims wherein some of the transducers (14) are energized at one frequency and other transducers (14) at a different frequency.

VSDOCID: <WO\_\_\_\_\_0035579A1\_I\_>



# INTERNATIONAL SEARCH REPORT

Inter anal Application No PCT/GB 99/03857

	OUT OF THE PARTY O		7 C17 GD 337 U3037	
IPC 7	SIFICATION OF SUBJECT MATTER B01J19/10			
According	to International Patent Classification (IPC) or to both national cla			
4	S SEARCHED	assification and IPC		
Minimum o	documentation searched (classification system followed by class	sification symbols)		
IPC 7	B01J			
Documenta	ation searched other than minimum documentation to the extent	that such documents are inclu	ded in the fields searched	
Electronic	data base consulted during the international search (name of da	ata base and, where practical,	search terms used)	
С. DOCUM	ENTS CONSIDERED TO BE RELEVANT			
Category '	Citation of document, with indication, where appropriate, of the	ne relevant passages	Relevant to claim No.	
X	US 5 658 534 A (DESBOROUGH COL	1,4		
	ROGER B, WARD LAWRENCE D) 19 August 1997 (1997-08-19)			
	the whole document			
v	CP 2 206 202 A (DDITTO) AND TO			
X	GB 2 306 202 A (BRITISH NUCLEA 30 April 1997 (1997-04-30)	R FUELS PLC)	1	
Α	the whole document		4	
,	US 4 550 467 4 (16)		7	
Α	US 4 556 467 A (KUHN MARTIN C, VERNON) 3 December 1985 (1985-	ZEITZ	5	
	claims 14,15	12-03)		
ĺ			]	
Furthe	er documents are listed in the continuation of box C.			
		X Patent family me	mbers are listed in annex.	
	egories of cited documents :	"T" later document publish	ed after the international filing date	
conside	nt defining the general state of the art which is not ared to be of particular relevance	or priority date and no cited to understand th	ot in conflict with the application but the principle or theory underlying the	
E" earlier do filing da	ocument but published on or after the international ite	invention "X" document of particular	relevance; the claimed invention	
Writer is	it which may throw doubts on priority claim(s) or a cited to establish the publication date of another	involve an inventive s	novel or cannot be considered to tep when the document is taken alone	
chanon e	or other special reason (as specified)  treferring to an oral disclosure, use, exhibition or	cannot be considered	relevance; the claimed invention to involve an inventive step when the	
otner me	eans  t published prior to the international filing date but	ments, such combination the art.	d with one or more other such docu- tion being obvious to a person skilled	
iater tha	in the phority date daimed	"&" document member of the	he same patent family	
Date of the ac	ctual completion of the international search	Date of mailing of the	international search report	
11	February 2000	21/02/2000		
lame and ma	tilling address of the ISA	Authorized officer		
	European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl.			
_	Fax: (+31-70) 340-3016	Vlassis,	M	

Form PCT/ISA/210 (second sheet) (July 1992)

## INTERNATIONAL SEARCH REPORT

Information on patent family members

Inter onal Application No PCT/GB 99/03857

Patent document cited in search report		Publication date	Patent family member(s)	Publication date	
US 5658534	А	19-08-1997	DE 69102918 D DE 69102918 T EP 0449008 A ES 2056510 T GB 2243092 A,B	25-08-1994 02-02-1995 02-10-1991 01-10-1994 23-10-1991	
GB 2306202	Α	30-04-1997	NONE		
US 4556467	Α	03-12-1985	NONE		

Form PCT/ISA/210 (patent family annex) (July 1992)